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Analytical solution for facilitated transport across a membrane

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Abstract

An analytical expression for the facilitation factor of component A across a liquid membrane is derived in case of an instantaneous reaction $A(g) + B(l) \Leftrightarrow AB(l)$ inside the liquid membrane. The present expression has been derived based on the analytical results of Olander (A.I.Ch.E. J. 6(2) (1960) 233) obtained for the enhancement factor for G–L systems with bulk. The analytical expression for the facilitation factor allows for arbitrary diffusivities of all species involved and does not contain any simplification or approximations. The facilitation factor starts from the value of unity, goes through a maximum and then reduces back to unity as the equilibrium constant is increased. The maximum facilitation factor occurs at higher values of the equilibrium constant as the ratio of the permeate-complex over carrier diffusivity is reduced whereas the maximum facilitation factor occurs at the same value of the equilibrium constant for all values of D_A/D_B (ratio of the permeate over carrier diffusivity). A similar behavior is seen for the flux of A as a function of the equilibrium constant. The facilitation factor remains constant with changes in the film thickness whereas the flux of A reduces with an increase in the thickness of the film. A linear increase of the facilitation factor and flux of A are seen with increasing initial carrier concentration.

Keywords: Facilitated transport; Diffusion coefficients; Liquid membranes; Instantaneous reaction; Enhancement factor

1. Introduction

Facilitated transport is a process in which a chemical carrier binds reversibly and selectively with permeate species at a feed side of a film, transports the permeate through the film and, then, releases it at the permeate side of the film. In this process, chemical reaction and diffusion occur simultaneously in the system which accelerates the transport of the permeate species through the film. The film may either be an immobilized liquid film (e.g. a liquid impregnated in a porous inert structure) in which all species can move freely and the porous structure is an inert or a membrane in which the porous structure has an active contribution in the facilitated transport (e.g. ion selective membranes). There are several articles that describe facilitated transport in detail (Ward, 1970; Schultz, Goddard, & Suchdeo, 1974a,b; Smith, Lander, & Quinn, 1977; Goddard, 1977; Way, Noble, Flynn, & Sloan, 1982).

Facilitated transport has received a great deal of attention because of its numerous promising applications. Examples of these applications are transport of O_2 through hemoglobin solutions (Scholander 1960) CO through films of cuprous chloride solutions (Smith and Quinn, 1980), CO_2 through liquid membranes for various amine solutions (Teramoto et al., 1997) CO_2 in ion exchange membranes for different ionomer films (Noble, Pellegrino, Grosgogeat, Sperry, & Way, 1988; Yamaguchi, Koval, & Noble, 1996), and CO_2 through amine/polymer solution (Yamaguchi, Boetje, Koval, Noble, & Bowman, 1995). Separation and transport of a CO_2/N_2 mixture through a liquid membrane of diethanolamine (Guha, Majumdar, & Sirkar, 1990), CO_2/CH_4 through a liquid membrane of amine solutions (Teramoto, Nakai, & Ohnishi, 1996) and removal of CH_4 from a ternary mixture of CO_2, H_2S and CH_4 (Way & Noble, 1989), and H_2S from CO_2 (Kreulen, Smolders, & Versteeg, 1993) are other examples of the possible applications. Some other applications include transport and separation of 1-hexane and 1,5-hexadiene through a film of

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a Nafion membrane (Koval & Spontarellit, 1988), ethene and ethane through Nafion membranes (Eriksen, Aksnes, & Dahl, 1993), olefin/paraffin mixture through polymer membranes (Ho & Dalrymple, 1994; VanZyl & Linkov, 1997), hydrocarbons mixture using ion exchange membranes (VanZyl, Kerres, & Cui, 1997) and styrene and ethylbenzene through ionomer membranes (Koval, Spontarelli, & Noble, 1989; Koval, Spontarelli, & Thoen, 1992).

2. Literature review for the reaction $A(g) + B(l) \Leftrightarrow AB(l)$

The most common generalized and overall reaction scheme for the transport of a gaseous component across a liquid film reported in the literature is

$$A(g) + B(l) \quad \Leftrightarrow \quad AB(l), \tag{1}$$

where A, B and AB are permeate species, mobile carrier, and permeate–carrier complex, respectively. Since the governing equations that describe the steady state transport of permeate species across the liquid film of simultaneous chemical reaction and diffusion of the above reaction are coupled and usually nonlinear, a generally applicable analytical solution for arbitrary kinetics is not possible.

There have been several attempts to obtain analytical solutions for such systems, however, most of the analytical approaches reported are only valid for simplified situations not completely representing the actual process in the membrane. An analytical solution for the steady state facilitation factor, which is defined as the ratio of the actual solute flux (with chemical reaction) to the solute diffusion flux only, was developed by Smith and Quinn (1979) in rectangular coordinates assuming equilibrium concentrations. The authors linearized the reaction rate expression by assuming that there is a large excess of mobile carrier. Smith and Quinn additionally assumed equal diffusivity of carrier and complex in their analysis. They reported the correct behavior for asymptotic reaction- and diffusion-limited cases at low equilibrium constant. However, the accuracy of the facilitation factor obtained by this approach decreases at large values of the equilibrium constant (refer to Fig. 1). Noble, Way, and Power (1986) derived an expression for the facilitation factor using the similar approach as Smith and Quinn which also accounts for external mass transfer resistance. The equation for the facilitation factor obtained by the authors reduces to Smith and Quinn's for the case of no external mass transfer. Also in this paper, the authors used equal diffusivity of the carrier and complex in the analysis. The accuracy of the facilitation factor obtained by the authors decreases at high equilibrium constant, similar to Smith and Quinn. An improved method for the evaluation of facilitation factor is reported by Jemaa and Noble (1992). The authors empirically determined a nonzero permeate concentration at the exit of the membrane by matching the facilitation factor resulting from Smith and Quinn's model with the one computed numerically by Kemena, Noble, and Kemp (1983). Again, in this improved method equal diffusivities of the carrier and complex were assumed. However, the introduction of an empirical fit parameter is not very elegant. Noble (1990) derived a model for the facilitation of neutral molecules such as O_2 across a fixed site carrier membrane. The analysis of the analytical determination of the facilitation factor is similar to the one presented by Smith and Quinn (1979) for equal carrier and complex diffusivities.

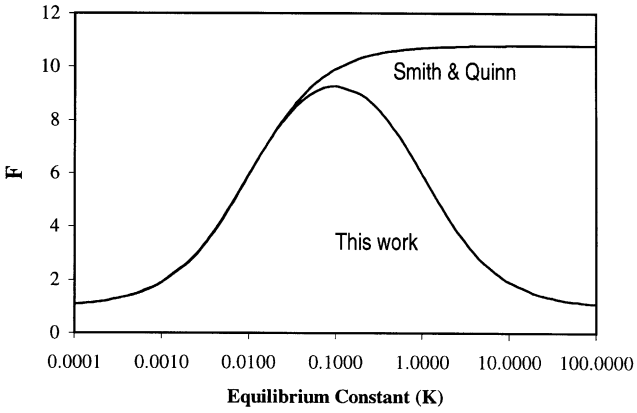

Fig. 1. Comparison of the effect of facilitation factor as a function of equilibrium constant between Smith and Quinn and the exact solution for $D_{AB} = D_A = 10^{-9}$ (m^2/s), $\alpha = 1$ (−), $C_{A0} = 100$ (mol/m^3), $C_{AL} = 1$ (mol/m^3), $C_{\text{Binitial}} = 1000$ (mol/m^3), and $L = 0.001$ (m).

Basaran, Burban, and Auvil (1989) presented two models for the facilitated transport of unequal carrier and permeate-carrier complex diffusivities which also permit arbitrary kinetic rates. The first method was derived for very low Damkohler numbers ($k_r L^2/D_A$) and solved analytically using regular perturbation analysis. This has limited industrial application because for low Damkohler number, the contribution of the carrier to facilitated transport is low. The second model was solved numerically using a Galerkin/finite element method that was solved by an iteration technique using Newton's method. Teramoto (1994) presented an approximate solution for the facilitation factor. Using trial & error calculations, approximate facilitation factors were calculated and compared with several reported numerical solutions with a good agreement. The model is valid for both equal and unequal diffusivities.

A model by Chaara and Noble (1989) describes diffusion, chemical reaction and convection across a liquid film. The authors obtained a facilitation factor for equal diffusivities of carrier and a complex by assuming equilibrium composition and a constant carrier concentration. This model is an extension of the expression obtained by Noble et al. (1986) accounting for the convective flow across the film, however, with the same limitation(s).

Beside these semi-analytical solutions, of course, several authors solved the governing equations that describe the steady state transport of permeate species across the liquid film of the above system numerically. Jain and Schultz (1982) solved the system of differential equations

using orthogonal collocation for both equal and unequal diffusivities. Kemena et al. (1983) studied the optimization of the facilitation factor for equal diffusivities of carrier and complex using a numerical technique. Kirkkoprudindi and Noble (1989) presented the numerical results obtained for multiple site transport using equal carrier and complex diffusivities. Dindi, Noble, and Koval (1992) solved the model for the competitive facilitated transport numerically for unequal diffusivities of carrier and complex. An equilibrium composition with a constant concentration of carrier was used. The equation obtained for the facilitation factor in this study reduced to the one of Smith and Quinn (1979) for one permeate and equal diffusivities of carrier and complex.

Unsteady state competitive facilitated transport of two gases through the membrane was determined numerically by Niiya and Noble (1985). This model is compared with other models for the case of steady state and "equilibrium core" with good agreement. Folkner and Noble (1983) reported the transient flux for one-dimensional transport in rectangular, cylindrical and spherical co-ordinates using numerical techniques.

Nearly all models mentioned in the previous section are either inaccurate (especially the approximate analytical solutions as the problem is oversimplified) or elaborate to work with (numerical models). Except for the solutions by Basaran et al. (1989) (partly numerical) and Teramoto (1994) (using an assumption on the concentration of B), all other analytical solutions have not been able to accurately predict the facilitation factor over the entire range from diffusion- to reaction-limited mass transport. Furthermore, all analytical analyses have been restricted to equal diffusivity of the mobile carrier and permeate-carrier complex except the analyses presented by Basaran et al. and Teramoto. It is quite clear that there is a need for a simple method to reliably predict the facilitation factor.

The aim of this paper is to derive an analytical equation for the facilitation factor that allows for unequal diffusivity of the carrier and permeate-carrier complex for reactions instantaneous with respect to mass transfer. The method is based on a simple equation derived by Olander (1960). Originally Olander derived equations for instantaneous reactions for systems with a liquid bulk, but the solution is also applicable to instantaneous reactions in liquid films. Previously, Kreulen et al. (1993) showed that the facilitation factor for the reaction $A + B \leftrightarrow C + D$ using equal diffusivities of all components could be calculated analytically using Olander's approach. It will be shown that the present method, based on Olander's equations, is applicable for instantaneous reactions with arbitrary values of the diffusivities of the various species.

3. Theory

The governing equations for steady state, one-dimensional transport in a rectangular geometry of the overall reaction scheme shown in Eq. (1) are as follows:

$$D_A \frac{d^2C_A}{dx^2} + k_1\left(-C_AC_B + \frac{C_{AB}}{K}\right) = 0, \tag{2}$$

$$D_B \frac{d^2C_B}{dx^2} + k_1\left(-C_AC_B + \frac{C_{AB}}{K}\right) = 0, \tag{3}$$

$$D_{AB} \frac{d^2C_{AB}}{dx^2} + k_1\left(C_AC_B - \frac{C_{AB}}{K}\right) = 0, \tag{4}$$

where C_A, C_B, C_{AB} are the concentration of permeate (A), mobile carrier (B), and permeate–carrier complex (AB), respectively, D_A, D_B, and D_{AB} are the diffusivity of A, B and AB, respectively, k_1 is the forward reaction rate, K is the equilibrium constant, and x is the distance.

The boundary conditions for Eqs. (2)–(4) are given as

$$x = 0, \quad C_A = C_{A0}, \quad \frac{dC_B}{dx} = \frac{dC_{AB}}{dx} = 0, \tag{5}$$

$$x = L, \quad C_A = C_{AL}, \quad \frac{dC_B}{dx} = \frac{dC_{AB}}{dx} = 0. \tag{6}$$

Eqs. (2)–(4) can be simplified to

$$D_A \frac{d^2C_A}{dx^2} + D_{AB} \frac{d^2C_{AB}}{dx^2} = 0, \tag{7}$$

$$D_B \frac{d^2C_B}{dx^2} + D_{AB} \frac{d^2C_{AB}}{dx^2} = 0. \tag{8}$$

For an instantaneous reaction

$$K = \frac{C_{AB}}{C_AC_B}. \tag{9}$$

The boundary conditions for an instantaneous reaction change to

$$x = 0, \quad C_A = C_{A0}, \quad D_B \frac{dC_B}{dx} + D_{AB} \frac{dC_{AB}}{dx} = 0,$$
$$C_{AB0} = KC_{A0}C_{B0}, \tag{10}$$

$$x = L, \quad C_A = C_{AL}, \quad D_B \frac{dC_B}{dx} + D_{AB} \frac{dC_{AB}}{dx} = 0,$$
$$C_{ABL} = KC_{AL}C_{BL}. \tag{11}$$

The problem is now identical to the problem of mass transfer with simultaneous reactions according to the film model for systems with a bulk. This model has been successfully solved by Olander (1960) not only for the present instantaneous reaction of $A(g)+B(l) \Leftrightarrow AB(l)$ but also for other instantaneous reactions including $A(g)+B(l) \Leftrightarrow C(l)+D(l)$. If the solution of Olander for the present reaction is considered, one can see that the concentration of B at $x = L$ is required. For a system with bulk the concentration of B is imposed by the liquid loading and the assumption of equilibrium in the liquid bulk. However, for the present case the

following condition has to be satisfied:

$$\int_0^L (C_B(x) + C_{AB}(x))\,\mathrm{d}x = C_{\mathrm{Binitial}} L. \tag{12}$$

It can be shown however that (refer to Appendix A):

$$C_B + \frac{D_{AB}}{D_B} C_{AB} = \frac{C^*}{D_B}. \tag{13}$$

This gives for the concentration of B at $x = L$:

$$C_{BL} = C_{B0} \frac{1 + \alpha K C_{A0}}{1 + \alpha K C_{AL}}, \tag{14}$$

where C_{B0} can be obtained from the equation below:

$$C_{B0} = \frac{C^*}{D_B(1 + \alpha K C_{A0})} \tag{15}$$

or

$$C_{BL} = \frac{C^*}{D_B(1 + \alpha K C_{AL})}. \tag{16}$$

In this equation C^* is defined according to implicit equation (B.28) or the identical expression (B.44) in Appendix B.

The facilitation factor can now be calculated to be

$$F = 1 + \frac{\alpha K C^*}{D_A(1 + (D_{AB}/D_B)K C_{AL})(1 + (D_{AB}/D_B)K C_{A0})} \tag{17}$$

and the flux according to

$$J_A = F \frac{D_A}{L}(C_{A0} - C_{AL}). \tag{18}$$

Eq. (17) starts at the value of 1, increases to its maximum value, and, then, reduces to the value of unity as the equilibrium constant is increased from a low to high value.

4. Results and discussion

Fig. 2 shows a typical relationship between the facilitation factor and equilibrium constant for different values of D_A/D_B, which is the ratio of the diffusivity of permeate to that of carrier. As expected, the facilitation factor starts at the value of unity, increases to its maximum value, and then drops back to unity as the value of the equilibrium constant is increased. As is presented in Fig. 2, the maximum occurs at the same value of the equilibrium constant for all values of D_A/D_B. However, the maximum value of the facilitation factor increases as the ratio of D_A/D_B reduces. The relationship between the facilitation factor and equilibrium constant for different values of α ($\alpha = D_{AB}/D_B$) is presented in Fig. 3a. Similar to Fig. 2, the facilitation factor starts at the value of 1, increases to its maximum value, and then drops back to unity as the value of the equilibrium constant is increased. The maximum value of the facilitation factor increases as the ratio of α reduces, however, the maximum facilitation factor occurs at a higher equilibrium constant as the value of α is reduced. The relationship between the flux of A and equilibrium constant for different values of α ($\alpha = D_{AB}/D_B$) is presented in Fig. 3b, which shows a similar behavior as Fig. 3a. Fig. 4a shows the relationship between the facilitation factor and equilibrium constant for different values of the initial permeate concentration (C_{A0}). As expected, the facilitation factor goes through a maximum for all values of C_{A0}, however, the maximum value of the facilitation factor increases as C_{A0} (or C_{A0}/C_{AL}) is reduced. In addition, this maximum occurs at higher values of the equilibrium constant as C_{A0} is reduced. The relationship between the flux of A and equilibrium constant for different

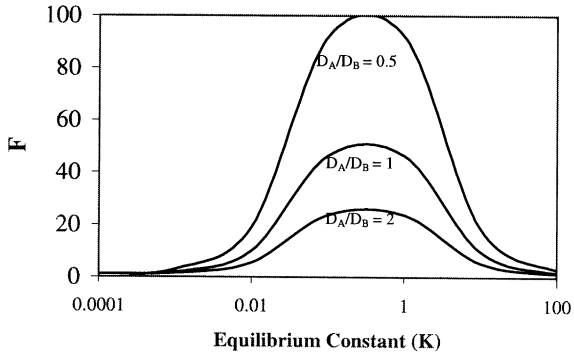

Fig. 2. Effect of equilibrium constant on facilitation factor for different values of D_A/D_B for the case of $D_B = D_{AB} = 10^{-9}$ (m^2/s), $L = 0.001$ (m), $C_{A0} = 10$ (mol/m^3), $C_{AL} = 1$ (mol/m^3), $C_{\mathrm{Binitial}} = 1000$ (mol/m^3).

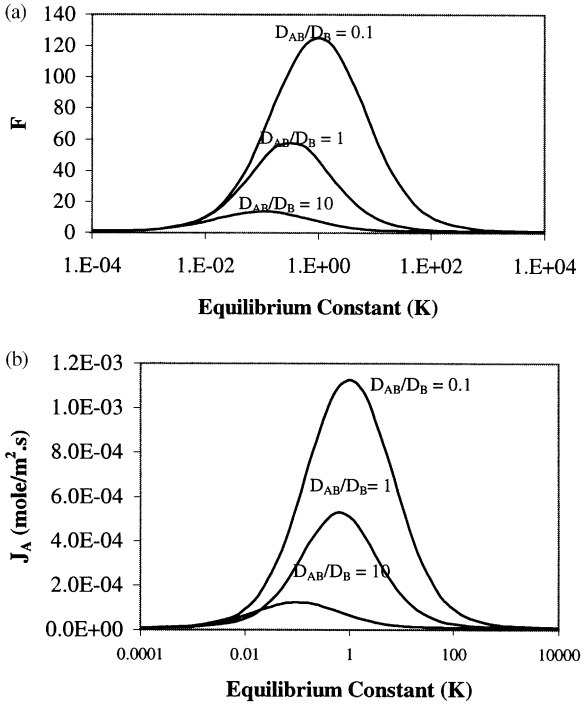

Fig. 3. (a) Effect of equilibrium constant on facilitation factor for different values of $\alpha(D_{AB}/D_B)$ for the case $D_{AB} = D_A = 10^{-9}$ (m^2/s), $L = 0.001$ (m), $C_{A0} = 10$ (mol/m^3), $C_{AL} = 1$ (mol/m^3), $C_{\mathrm{Binitial}} = 1000$ (mol/m^3). (b) Effect of equilibrium constant on flux of A for different values of $\alpha(D_{AB}/D_B)$ for the case $D_{AB} = D_A = 10^{-9}$ (m^2/s), $L = 0.001$ (m), $C_{A0} = 10$ (mol/m^3), $C_{AL} = 1$ (mol/m^3), $C_{\mathrm{Binitial}} = 1000$ (mol/m^3).

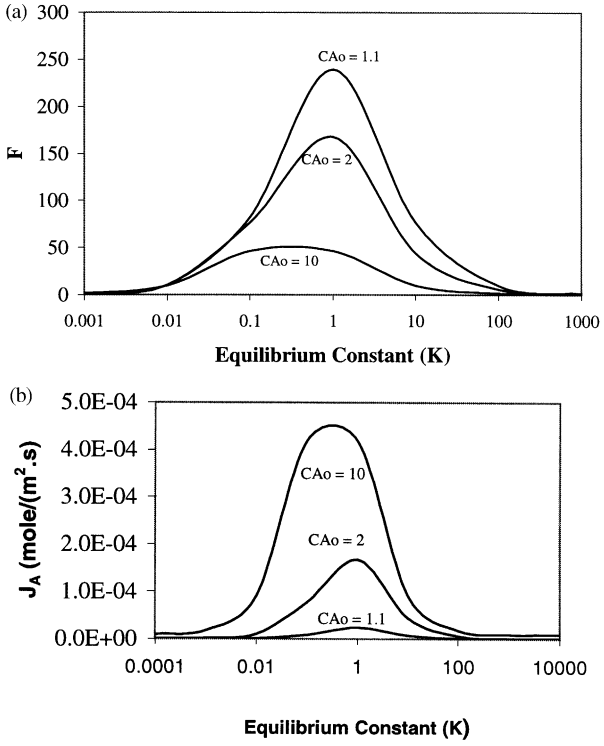

Fig. 4. (a) Effect of equilibrium constant on facilitation factor for different values of initial permeate concentration for the case of $D_{AB} = D_A = D_B = 10^{-9}$ (m^2/s), $L = 0.001$ (m), $C_{AL} = 1$ (mol/m^3), $C_{\text{Binitial}} = 1000$ (mol/m^3). (b) Effect of equilibrium constant on flux of A for different values of initial permeate concentration for the case of $D_{AB} = D_A = D_B = 10^{-9}$ (m^2/s), $L = 0.001$ (m), $C_{AL} = 1$ (mol/m^3), $C_{\text{Binitial}} = 1000$ (mol/m^3).

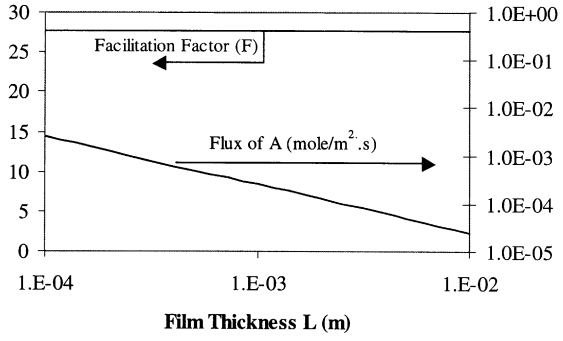

Fig. 5. Facilitation factor and flux of A as a function of film thickness for the case of $D_{AB} = D_A = 10^{-9}$ (m^2/s), $D_B = 5 \times 10^{-10}$ (m^2/s), $K = 1$ (m^3/mol), $C_{A0} = 10$ (mol/m^3), $C_{AL} = 1$ (mol/m^3), $C_{\text{Binitial}} = 1000$ (mol/m^3).

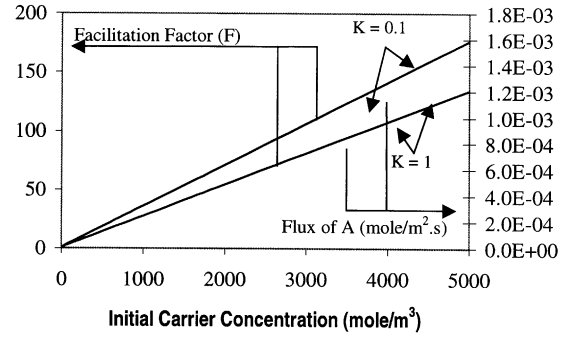

Fig. 6. Facilitation factor and flux of A as a function of initial carrier concentration for different values of equilibrium constant for the case of $D_{AB} = D_A = 10^{-9}$ (m^2/s), $D_B = 5 \times 10^{-10}$ (m^2/s), $L = 0.001$ (m), $C_{A0} = 10$ (mol/m^3), $C_{AL} = 1$ (mol/m^3), $C_{\text{Binitial}} = 1000$ (mol/m^3).

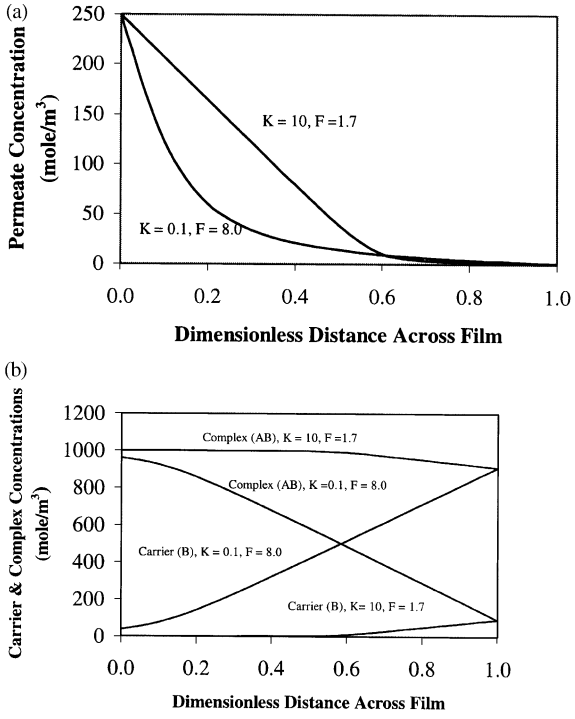

Fig. 7. Permeate concentration profile for different values of equilibrium constants for the case of $D_{AB} = D_B = 10^{-9}$ (m^2/s), $D_A = 5 \times 10^{-10}$ (m^2/s), $L = 0.001$ (m), $C_{A0} = 250$ (mol/m^3), $C_{AL} = 1$ (mol/m^3), $C_{\text{Binitial}} = 1000$ (mol/m^3), F corresponds to facilitation factor. (b) Carrier and complex concentration profile for different values of equilibrium constants for the case of $D_{AB} = D_B = 10^{-9}$ (m^2/s), $D_a = 5 \times 10^{-10}$ (m^2/s), $L = 0.001$ (m), $C_{A0} = 250$ (mol/m^3), $C_{AL} = 1$ (mol/m^3), $C_{\text{Binitial}} = 1000$ (mol/m^3), F corresponds to facilitation factor.

values of initial permeate concentration (C_{A0}) is presented in Fig. 4b. The flux of A goes through a maximum for all values of C_{A0}, however, the maximum value of the flux of A increases as C_{A0} (or C_{A0}/C_{AL}) is increased. In addition, this maximum occurs at higher values of the equilibrium constant as C_{A0} is reduced. Fig. 5 shows the relationship between the facilitation factor and flux of A as a function of membrane thickness. The facilitation factor remains constant as the membrane thickness is increased, whereas, the flux of A drops with the membrane thickness linearly. As presented in the last part of Appendix B, C^* is independent of the thickness, causing the facilitation factor to remain unchanged with the thickness of the film. The relationship between the facilitation factor and flux of A as a function of the initial carrier concentration for different values of the equilibrium constant is presented in Fig. 6. Both the facilitation factor and flux of A increase linearly with increasing initial carrier concentration. Fig. 7a shows the permeate

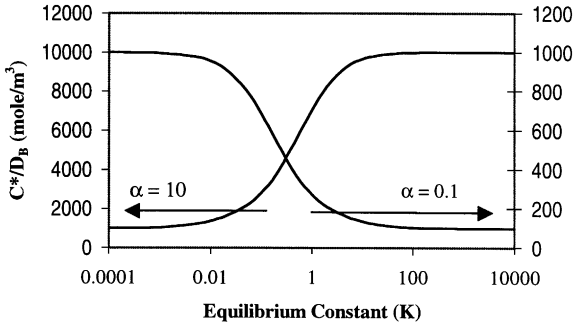

Fig. 8. Effect of equilibrium constant on C^*/D_b for different values of $\alpha\,(D_{AB}/D_B)$ for the case of $D_{AB} = D_A = 10^{-9}$ (m^2/s), $L = 0.001$ (m), $C_{A0} = 10$ (mol/m^3), $C_{AL} = 1$ (mol/m^3), $C_{Binitial} = 1000$ (mol/m^3).

concentration profile for different values of the equilibrium constant. The permeate concentration is at its maximum at the high-pressure side of the membrane and then drops across the membrane until it reaches the permeate concentration at low-pressure side of the membrane. Carrier and complex concentration profiles for the corresponding values of the equilibrium constant are presented in Fig. 7b. The concentration of the carrier at the high-pressure side of the film is initially low and increases along the thickness of the film, whereas, the permeate–carrier concentration is high at $x = 0$ and then decreases to its minimum value at the low-pressure side of the film ($x = L$). The relationship between C^*/D_B and equilibrium constant for different values of α ($\alpha = D_{AB}/D_B$) is presented in Fig. 8. The value of C^*/D_B starts at $C_{Binitial}$ for low equilibrium constants and then it goes through a S-shaped curve and finally converges to the value of $\alpha C_{Binitial}$ as the equilibrium constant is increased. It is also important to note that, for $\alpha = 1$, C^*/D_B will remain constant and it has the same value of $C_{Binitial}$ for all values of the equilibrium constant.

The present contribution deals with the reaction $A(g) + B(l) \Leftrightarrow AB(l)$ as it is the most frequently encountered from of facilitated transport. However, for other reactions the appropriate solution of Olander (1960), derived for G–L systems with bulk) can also be used in a similar way to derive the accompanying (analytical) expression for the facilitation factor across a membrane.

5. Conclusion

An analytical equation for the facilitation factor, which allows for unequal diffusivity of the carrier and permeate–carrier complex for reactions instantaneous with respect to mass transfer was derived for the system of $A(g) + B(l) \Leftrightarrow AB(l)$. The present analytical solution has been derived using the results of Olander (1960) for instantaneous reactions in G–L systems with bulk. The analytical solution can be applied for any set of diffusivities of reactants and product and does not contain any approximation or simplification.

The facilitation factor starts from the value of unity, goes through a maximum and then reduces back to unity as the equilibrium constant is increased with the changes in diffusion coefficients of A, B and/or AB, and with the changes in initial permeate concentrations. The maximum facilitation factor occurs at higher values of the equilibrium constant as the ratio of the permeate-complex over carrier diffusivity is reduced whereas the maximum facilitation factor occurs at the same value of equilibrium constant for all values of D_A/D_B (ratio of the permeate over carrier diffusivity). A similar behavior is seen for the flux of A as a function of equilibrium constant. Facilitation factor remains constant with changes in the film thickness whereas the flux of A reduces with an increase in the thickness of the film. A linear increase on facilitation factor and flux of A are seen with increasing initial carrier concentration. The values of C^*/D_B starts at $C_{Binitial}$ for low equilibrium constants and then it goes through a S-shaped curve and finally converges at the value of $\alpha C_{Binitial}$ as the equilibrium constant is increased.

Notation

C_A	concentration of permeate A, mol/m^3
C_{A0}	initial concentration of permeate A, mol/m^3
C_{AL}	final concentration of permeate A, mol/m^3
C_B	concentration of mobile carrier B, mol/m^3
C_{AB}	concentration of permeate-carrier complex AB, mol/m^3
$C_{Binitial}$	initial concentration of carrier, mol/m^3
D_A	diffusion coefficient of permeate A, m^2/s
D_B	diffusion coefficient of mobile carrier B, m^2/s
D_{AB}	diffusion coefficient of permeate-carrier complex AB, m^2/s
F	facilitation factor, dimensionless
J_A	flux of A across membrane, mol/m^2s
k_1	forward reaction rate constant, $m^3/mol\ s$
K	equilibrium constant, m^3/mol
L	characteristic length or thickness of membrane, m
x	position, m
α	ratio of the carrier and complex diffusivities, dimensionless

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Appendix A. Derivation of expression for the facilitation factor

The governing equations are:

$$D_A \frac{d^2C_A}{dx^2} + k_1\left(-C_AC_B + \frac{C_{AB}}{K}\right) = 0, \tag{A.1}$$

$$D_B \frac{d^2C_B}{dx^2} + k_1\left(-C_AC_B + \frac{C_{AB}}{K}\right) = 0, \tag{A.2}$$

$$D_{AB} \frac{d^2C_{AB}}{dx^2} + k_1\left(C_AC_B - \frac{C_{AB}}{K}\right) = 0. \tag{A.3}$$

The boundary conditions for Eqs. (A.1)–(A.3) are given as

$$x = 0, \quad C_A = C_{A0}, \quad \frac{dC_B}{dx} = \frac{dC_{AB}}{dx} = 0, \tag{A.4}$$

$$x = L, \quad C_A = C_{AL}, \quad \frac{dC_B}{dx} = \frac{dC_{AB}}{dx} = 0. \tag{A.5}$$

This system is simplified by addition of Eqs. (A.1) and (A.3) and (A.2) and (A.3), the resultant equations are:

$$D_A \frac{d^2C_A}{dx^2} + D_{AB} \frac{d^2C_{AB}}{dx^2} = 0, \tag{A.6}$$

$$D_B \frac{d^2C_B}{dx^2} + D_{AB} \frac{d^2C_{AB}}{dx^2} = 0. \tag{A.7}$$

For instantaneous reaction

$$K = \frac{C_{AB}}{C_AC_B}. \tag{A.8}$$

Eqs. (A.6)–(A.8) can be solved analytically using the appropriate boundary conditions. The boundary conditions for an instantaneous reaction are:

$$x = 0, \quad C_A = C_{A0}, \tag{A.9a}$$

$$x = L, \quad C_A = C_{AL}. \tag{A.9b}$$

$$x = 0 \,\&\, x = L, \quad D_B \frac{dC_B}{dx} + D_{AB} \frac{dC_{AB}}{dx} = 0, \tag{A.10}$$

Solutions to Eqs. (A.6) and (A.7) are:

$$D_AC_A + D_{AB}C_{AB} = a_1x + a_2, \tag{A.11}$$

$$D_BC_B + D_{AB}C_{AB} = a_3x + a_4, \tag{A.12}$$

where a_1, a_2, a_3 and a_4 are the constants that will be determined.

Differentiating Eq. (A.12) and applying Eq. (A.9b) results in a_3 to be 0. Therefore, Eq. (A.12) reduces to

$$D_BC_B + D_{AB}C_{AB} = a_4 \tag{A.13}$$

or

$$D_BC_B + D_{AB}KC_AC_B = a_4. \tag{A.14}$$

Solving for C_B:

$$C_B = \frac{a_4}{D_B(1 + \alpha KC_A)}, \tag{A.15}$$

where

$$\alpha = \frac{D_{AB}}{D_B}.$$

From Eqs. (A.11) and (A.8):

$$D_AC_A + D_{AB}KC_AC_B = a_1x + a_2. \tag{A.16}$$

Combination of Eqs. (A.16) and (A.15) leads to

$$D_AC_A + \alpha \frac{KC_A}{(1 + \alpha KC_A)} a_4 = a_1x + a_2, \tag{A.17}$$

where a_1, a_2 and a_4 need to be determined.

Using boundary condition (A.9a), Eq. (A.17) results in:

$$D_AC_{A0} + \alpha \frac{KC_{A0}}{(1 + \alpha KC_{A0})} a_4 = a_2 \tag{A.18}$$

and using boundary condition (A.10), Eq. (A.17) results in:

$$D_AC_{AL} + \alpha \frac{KC_{AL}}{(1 + \alpha KC_{AL})} a_4 = a_1L + a_2. \tag{A.19}$$

Combining Eqs. (A.18) and (A.19), and solving for a_1:

$$a_1 = \frac{D_A(C_{AL} - C_{A0}) + \alpha Ka_4(C_{AL}/(1 + \alpha KC_{AL}) - C_{A0}/(1 + \alpha KC_{A0}))}{L}. \tag{A.20}$$

To determine a_4: Eq. (A.15) is evaluated at $x = 0$ and $x = L$:

At $x = 0$,

$$C_{B0} = \frac{a_4}{D_B(1 + \alpha KC_{A0})}, \tag{A.21}$$

At $x = L$,

$$C_{BL} = \frac{a_4}{D_B(1 + \alpha KC_{AL})}, \tag{A.22}$$

where C_{B0} and C_{BL} need to be determined. Solving for a_4 and equating Eqs. (A.21) and (A.22) leads to

$$C_{BL} = C_{B0} \frac{1 + \alpha KC_{A0}}{1 + \alpha KC_{AL}}. \tag{A.23}$$

Therefore

$$C_{BL} + \alpha C_{ABL} = C_{BL} + \alpha KC_{AL}C_{BL} = C_{BL}(1 + \alpha KC_{AL}). \tag{A.24}$$

Combining Eqs. (A.23) with (A.24):

$$C_{BL} + \alpha C_{ABL} = C_{B0}\frac{1+\alpha KC_{A0}}{1+\alpha KC_{AL}}(1+\alpha KC_{AL}) = C_{B0}(1+\alpha KC_{A0}) = C_{B0} + \alpha KC_{A0}C_{B0} = C_{B0} + \alpha C_{AB0}.$$

Or

$$C_B + \alpha C_{AB} = \text{constant} = \frac{a_4}{D_B} = \frac{C^*}{D_B}. \quad (A.25)$$

From Eq. (A.25):

$$a_4 = C^*. \quad (A.26)$$

Eq. (A.20) can be simplified to

$$a_1 = \frac{D_A(C_{AL} - C_{A0}) + \alpha KC^*[(C_{AL} - C_{A0})/((1+\alpha KC_{AL})(1+\alpha KC_{A0}))]}{L} \quad (A.27)$$

To calculate the concentration gradient of A, B and AB the following steps are taken:
From Eqs. (A.17) and (A.26):

$$D_A C_A + \alpha\frac{KC_A}{(1+\alpha KC_A)}C^* = a_1 x + a_2, \quad (A.28)$$

where a_1 is given by Eq. (A.27) and a_2 can be evaluated by combining Eqs. (A.18) and (A.26):

$$a_2 = D_A C_{A0} + \alpha\frac{KC_{A0}}{(1+\alpha KC_{A0})}C^*. \quad (A.29)$$

Eq. (A.28) with the constants a_1 and a_2 (Eqs. (A.27) and (A.29)) is a quadratic equation, which can be solved for the concentration of $A(C_A)$ in the film.

$$C_A = \frac{-b + \sqrt{b^2 - 4ac}}{2a}, \quad (A.30)$$

where

$$a = \alpha KD_A,$$

$$b = D_A + \alpha KC^* - \alpha Ka_1 x - a_2\alpha K,$$

$$c = -(a_1 x + a_2).$$

The concentrations of $B(C_B)$ and $AB(C_{AB})$ in the film can be obtained taking the following steps:
From Eqs. (A.22) and (A.26):

$$C_B = \frac{C^*}{D_B(1+\alpha KC_A)}. \quad (A.31)$$

Note that Eq. (A.22) is valid for any point in the film and C_{AB} can be calculated using the following equations:
From Eq. (A.25):

$$C_{AB} = \frac{(C^*/D_B) - C_B}{\alpha}. \quad (A.32)$$

From Eq. (A.8):

$$C_{AB} = KC_A C_B. \quad (A.33)$$

The facilitation factor can now be calculated to be

$$F = \frac{-a_1}{-D_A((C_{AL} - C_{A0})/L)}. \quad (A.34)$$

Combination of Eqs. (A.27) and (A.34) and simplification leads to

$$F = 1 + \frac{\alpha KC^*}{D_A(1+(D_{AB}/D_B)KC_{AL})(1+(D_{AB}/D_B)KC_{A0})}. \quad (A.35)$$

Eq. (A.35) starts at the value of unity, increases to its maximum value, and then decreases to unity as K is increased. The expression for C^* is derived in Appendix B.

Appendix B. Derivation of expression for C^*

Use

$$\int_0^L (C_B + C_{AB})\,dx = C_{\text{Binitial}}L. \quad (B.1)$$

We know (from Eq. (A.15))

$$D_B C_B + D_{AB}C_{AB} = C^*. \quad (B.2)$$

Solving for C_{AB}:

$$C_{AB} = \frac{C^* - D_B C_B}{D_{AB}} = \frac{C^*}{D_{AB}} - \frac{C_B}{\alpha} \quad \text{where } \alpha = \frac{D_{AB}}{D_B}. \quad (B.3)$$

Substituting Eq. (B.3) into Eq. (B.1):

$$\int_0^L \left(C_B + \frac{C^*}{D_{AB}} - \frac{C_B}{\alpha}\right)dx = C_{\text{Binitial}}L. \quad (B.4)$$

Integrating Eq. (B.4):

$$\frac{C^*}{D_{AB}}L + \left(1 - \frac{1}{\alpha}\right)\int_0^L C_B\,dx = C_{\text{Binitial}}L. \quad (B.5)$$

Now, lets work with

$$\int_0^L C_B\,dx.$$

From Eq. (B.2) and the equilibrium concentration ($C_{AB} = KC_A C_B$):

$$C_B + \alpha KC_A C_B = \frac{C^*}{D_B}, \quad (B.6)$$

or

$$C_B = \frac{C^*}{D_B(1+\alpha K C_A)}. \tag{B.7}$$

Thus

$$\int_0^L C_B\,\mathrm{d}x = \int_0^L \frac{C^*}{D_B(1+\alpha K C_A)}\,\mathrm{d}x = \frac{C^*}{D_B}\int_0^L \frac{\mathrm{d}x}{1+\alpha K C_A}, \tag{B.8}$$

$$\frac{C^*}{D_B}\int_0^L \frac{\mathrm{d}x}{1+\alpha K C_A} = \frac{C^*}{D_B}\int_0^L \frac{\mathrm{d}x}{1+\alpha K(-b+\sqrt{b^2-4ac})/2a}, \tag{B.9}$$

where C_A, a, b and c are given as (refer to Appendix A)

$$C_A = \frac{-b+\sqrt{b^2-4ac}}{2a}, \tag{A.30}$$

$$a = \alpha K D_A,$$

$$b = D_A + \alpha K C^* - \alpha K a_1 x - a_2 \alpha K = A - Bx,$$

$$A = D_A + \alpha K C^* - a_2 \alpha K \quad \text{and} \quad B = \alpha K a_1,$$

$$c = -(a_1 x + a_2) = -a_1 x - a_2.$$

Eq. (B.9) can be written as

$$\frac{C^*}{D_B}\int_0^L \frac{2a\,\mathrm{d}x}{2a+\alpha K[-A+Bx+\sqrt{(A-Bx)^2+4aa_1x+4aa_2}]} \tag{B.10}$$

$$= \frac{2aC^*}{D_B}\int_0^L \frac{\mathrm{d}x}{2a-\alpha KA+\alpha KBx+\alpha K[\sqrt{A^2-2ABx+B^2x^2+4aa_1x+4aa_2}]} \tag{B.11}$$

$$= \frac{2aC^*}{D_B}\int_0^L \frac{\mathrm{d}x}{A_1+A_2x+A_3[\sqrt{B^2x^2+A_4x+A_5}]} \tag{B.12}$$

$$= \frac{2aC^*}{D_B}\int_0^L \frac{\mathrm{d}x}{A_1+A_2x+A_3B[\sqrt{x^2+(A_4/B^2)x+(A_5/B^2)}]} \tag{B.13}$$

$$= \frac{2aC^*}{D_B}\int_0^L \frac{\mathrm{d}x}{A_1+A_2x+A_6[\sqrt{x^2+A_7x+A_8}]} \tag{B.14}$$

$$= \frac{2aC^*}{D_B}\int_0^L \frac{\mathrm{d}x}{A_2(x+(A_1/A_2))+A_6[\sqrt{x^2+A_7x+A_8}]} \tag{B.15}$$

$$= A_{10}\int_0^L \frac{\mathrm{d}x}{A_2(x+A_9)+A_6[\sqrt{x^2+A_7x+A_8}]}, \tag{B.16}$$

where

$$A_1 = 2a - \alpha KA,$$

$$A_2 = \alpha KB = (\alpha K)^2 a_1,$$

$$A_3 = \alpha K,$$

$$A_4 = 4aa_1 - 2AB,$$

$$A_5 = 4aa_2 + A^2,$$

$$A_6 = A_3 B = \alpha KB = A_2,$$

$$A_7 = \frac{A_4}{B^2} = \frac{4aa_1-2AB}{B^2} = \frac{2(D_A-\alpha KC^*+\alpha Ka_2)}{\alpha Ka_1} = 2A_9,$$

$$A_8 = \frac{A_5}{B^2} = \frac{4aa_2+A^2}{B^2} = \frac{4\alpha KD_Aa_2+(D_A+\alpha KC^*-a_2\alpha K)^2}{(\alpha Ka_1)^2},$$

$$A_9 = \frac{A_1}{A_2} = \frac{2a-\alpha KA}{\alpha KB} = \frac{2\alpha KD_A-\alpha K(D_A+\alpha KC^*-\alpha Ka_2)}{(\alpha K)^2 a_1} = \frac{D_A-\alpha KC^*+\alpha Ka_2}{\alpha Ka_1},$$

$$A_{10} = \frac{2aC^*}{D_B} = \frac{2\alpha KD_AC^*}{D_B}.$$

Continuing with Eq. (B.16)):

$$= A_{10}\int_0^L \frac{\mathrm{d}x}{A_2[(x+A_9)+\sqrt{x^2+2A_9x+A_8}]} \tag{B.17}$$

$$= \frac{A_{10}}{A_2}\int_0^L \frac{\mathrm{d}x}{(x+A_9)+\sqrt{x^2+2A_9x+A_8}}. \tag{B.18}$$

Multiplying both the numerator and denominator by the conjugate:

$$\frac{A_{10}}{A_2}\int_0^L \frac{\lfloor (x+A_9)-\sqrt{x^2+2A_9x+A_8}\rfloor\,\mathrm{d}x}{[(x+A_9)+\sqrt{x^2+2A_9x+A_8}][(x+A_9)-\sqrt{x^2+2A_9x+A_8}]} \tag{B.19}$$

$$=\frac{A_{10}}{A_2}\int_0^L \frac{\lfloor (x+A_9)-\sqrt{x^2+2A_9x+A_8}\rfloor\,\mathrm{d}x}{(x+A_9)^2-(x^2+2A_9x+A_8)} \tag{B.20}$$

$$=\frac{A_{10}}{A_2}\int_0^L \frac{\lfloor (x+A_9)-\sqrt{x^2+2A_9x+A_8}\rfloor\,\mathrm{d}x}{A_9^2-A_8} \tag{B.21}$$

$$=\frac{A_{10}}{A_2(A_9^2-A_8)}\int_0^L [A_9+x-\sqrt{x^2+2A_9x+A_8}]\,\mathrm{d}x \tag{B.22}$$

$$=\frac{A_{10}}{A_2(A_9^2-A_8)}\left[A_9L+\frac{L^2}{2}-\int_0^L \sqrt{(x^2+2A_9x+A_8)}\,\mathrm{d}x\right] \tag{B.23}$$

$$=\frac{A_{10}}{A_2(A_9^2-A8)}\left[A_9L+\frac{L^2}{2}-\int_0^L \sqrt{(x^2+2A_9x+A_9^2)-(A_9^2-A_8)}\,\mathrm{d}x\right] \tag{B.24}$$

$$=\frac{A_{10}}{A_2(A_9^2-A_8)}\left[A_9L+\frac{L^2}{2}-\int_0^L \sqrt{(x+A_9)^2-(A_9^2-A_8)}\,\mathrm{d}x\right] \tag{B.25}$$

$$=\frac{A_{10}}{A_2(A_9^2-A_8)}\left[\begin{array}{l} A_9L+\dfrac{L^2}{2}-\left(\dfrac{x+A_9}{2}\sqrt{(x+A_9)^2-(A_9^2-A_8)}\right.\\ \quad -\dfrac{A_9^2-A_8}{2}\ln|x+A_9\\ \quad \left.+\sqrt{(x+A_9)^2-(A_9^2-A_8)}|\right)_0^L \end{array}\right] \tag{B.26}$$

$$=\frac{A_{10}}{A_2(A_9^2-A_8)}\left[\begin{array}{l} A_9L+\dfrac{L^2}{2}-\left(\dfrac{L+A_9}{2}\sqrt{(L+A_9)^2-(A_9^2-A_8)}\right.\\ \quad -\dfrac{A_9^2-A_8}{2}\ln\left|\begin{array}{l} L+A_9\\ \quad +\sqrt{(L+A_9)^2-(A_9^2-A_8)}\end{array}\right|\Bigg)\\ \quad +\left(\dfrac{A_9}{2}\sqrt{A_8.}-\dfrac{A_9^2-A_8}{2}\ln|A_9+\sqrt{A_8.}|\right) \end{array}\right], \tag{B.27}$$

With Eq. (B.5), the following expression can now be obtained:

$$\frac{C^*}{D_{AB}}L+\left(1-\frac{1}{\alpha}\right)\times\left(\frac{A_{10}}{A_2(A_9^2-A_8)}\left[\begin{array}{l}A_9L+\dfrac{L^2}{2}-\left(\dfrac{L+A_9}{2}\sqrt{(L+A_9)^2-(A_9^2-A_8)}\right.\\ \quad\left.-\dfrac{A_9^2-A_8}{2}\ln\left|\begin{array}{l}L+A_9\\ \quad+\sqrt{(L+A_9)^2-(A_9^2-A_8)}\end{array}\right|\right)\\ \quad+\left(\dfrac{A_9}{2}\sqrt{A_8.}-\dfrac{A_9^2-A_8}{2}\ln|A_9+\sqrt{A_8.}|\right)\end{array}\right]\right)=C_{\text{Binitial}}L. \tag{B.28}$$

After further simplification:

$$A_9^2-A_8=-\frac{4D_AC^*}{\alpha Ka_1^2}, \tag{B.29}$$

$$\frac{A_{10}}{A_2(A_9^2-A_8)}=-\frac{1}{2}\frac{a_1}{D_B}, \tag{B.30}$$

$$\begin{aligned}A_8&=\frac{A_5}{B^2}=\frac{4aa_2+A^2}{B^2}\\&=\frac{4\alpha KD_Aa_2+(D_A+\alpha KC^*-a_2\alpha K)^2}{(\alpha Ka_1)^2},\end{aligned} \tag{B.31}$$

$$\begin{aligned}A_9&=\frac{A_1}{A_2}=\frac{2a-\alpha KA}{\alpha KB}\\&=\frac{2\alpha KD_A-\alpha K(D_A+\alpha KC^*-\alpha Ka_2)}{(\alpha K)^2a_1}\\&=\frac{D_A-\alpha KC^*+\alpha Ka_2}{\alpha Ka_1},\end{aligned} \tag{B.32}$$

$$a_1=\frac{D_A(C_{AL}-C_{A0})+\alpha KC^*[(C_{AL}-C_{A0})/((1+\alpha KC_{AL})(1+\alpha KC_{A0}))]}{L} \tag{A.27}$$

$$a_2=D_AC_{A0}+\alpha\frac{KC_{A0}}{(1+\alpha KC_{A0})}C^*. \tag{A.29}$$

Eqs. (B.28)–(B.32) with Eqs. (A.27) and (A.29) give the relationship between constant C^* and C_{Binitial}. The equation is nonlinear and requires an iterative solution procedure.

B.1. Dependency of C^* on L (refer to Fig. 5)

To determine the dependency of C^* on the thickness of the film (L), the following steps are taken:

$$a_1=\frac{D_A(C_{AL}-C_{A0})+\alpha KC^*[(C_{AL}-C_{A0})/((1+\alpha KC_{AL})(1+\alpha KC_{A0}))]}{L}=\frac{C1}{L}, \tag{B.33}$$

$$a_2=D_AC_{A0}+\alpha\frac{KC_{A0}}{(1+\alpha KC_{A0})}C^*=C2, \tag{B.34}$$

$$A_9=\frac{D_A-\alpha KC^*+\alpha Ka_2}{\alpha Ka_1}=\frac{C3}{a_1}=L\frac{C3}{C1}=LC4, \tag{B.35}$$

$$\begin{aligned}A_8&=\frac{4\alpha KD_Aa_2+(D_A+\alpha KC^*-a_2\alpha K)^2}{(\alpha Ka_1)^2}\\&=\frac{C5}{a_1^2}=L^2\frac{C5}{C1^2}=L^2C6,\end{aligned} \tag{B.36}$$

$$A_9^2-A_8=-\frac{4D_AD_BC^*}{\alpha Ka_1^2}=\frac{C7}{a_1^2}=L^2\frac{C7}{C1^2}=L^2C8, \tag{B.37}$$

$$\frac{A_{10}}{A_2(A_9^2-A_8)}=-\frac{1}{2}\frac{a_1}{D_B}=a_1C9=\frac{C1C9}{L}=\frac{C10}{L}, \tag{B.38}$$

$$\begin{aligned}\sqrt{(L+A_9)^2-(A_9^2-A_8)}&=\sqrt{(L+L.C4)^2-L^2.C8}\\&=\sqrt{L^2[(1+C4)^2-C8]}\\&=L.\sqrt{[(1+C4)^2-C8]}\\&=L.C11,\end{aligned} \tag{B.39}$$

$$\ln \frac{(L + A_9 + \sqrt{(L + A_9)^2 - (A_9^2 - A_8)})}{A_9 + \sqrt{A_8}}$$
$$= \ln \frac{L + L.C4 + L.C11}{L.C4 + \sqrt{L^2.C6}}$$
$$= \ln \frac{L(1 + C4 + C11)}{L(C4 + \sqrt{C6})} = \ln \frac{C12}{C13} = C14. \quad \text{(B.40)}$$

Rearranging Eq. (B.28):

$$\frac{C^*}{D_{AB}} L + \left(1 - \frac{1}{\alpha}\right) \left(\frac{C10}{L}\right) \left(L^2 C4 + \frac{L^2}{2} - \frac{L + LC4}{2} LC11 + \frac{LC4}{2} \sqrt{L^2 C6} + \frac{L^2 C8}{2} C14\right) = C_{\text{Binitial}} L. \quad \text{(B.41)}$$

After further simplification, Eq. (B.41) reduces to

$$\frac{C^*}{D_{AB}} L + \left(1 - \frac{1}{\alpha}\right) \left(\frac{C10}{L}\right) L^2 \left(C4 + \frac{1}{2} - \frac{1 + C4}{2} C11 + \frac{C4}{2} \sqrt{C6} + \frac{C8}{2} C14\right) = C_{\text{Binitial}} L \quad \text{(B.42)}$$

or

$$\frac{C^*}{D_{AB}} L + L.C15 = C_{\text{Binitial}} L, \quad \text{(B.43)}$$

$$\frac{C^*}{D_{AB}} + C15 = C_{\text{Binitial}}, \quad \text{(B.44)}$$

where

$$C1 = D_A (C_{AL} - C_{A0}) + \alpha K C^* \left[\frac{C_{AL} - C_{A0}}{(1 + \alpha K C_{AL})(1 + \alpha K C_{A0})}\right],$$

$$C2 = D_A C_{A0} + \alpha \frac{K C_{A0}}{(1 + \alpha K C_{A0})} C^* = a_2,$$

$$C3 = \frac{D_A - \alpha K C^* + \alpha K a_2}{\alpha K},$$

$$C4 = \frac{C3}{C1}, \quad C5 = \frac{4\alpha K D_A a_2 + (D_A + \alpha K C^* - a_2 \alpha K)^2}{(\alpha K)^2},$$

$$C6 = \frac{C5}{C1^2}, \quad C7 = -\frac{4 D_A D_B C^*}{\alpha K},$$

$$C8 = \frac{C7}{C1^2}, \quad C9 = -\frac{1}{2 D_B},$$

$$C10 = C1.C9, \quad C11 = \sqrt{(1 + C4)^2 - C8},$$

$$C12 = 1 + C4 + C11, \quad C13 = C4 + \sqrt{C6},$$

$$C14 = \ln \frac{C12}{C13},$$

$$C15 = C10 \left(1 - \frac{1}{\alpha}\right) \left(C4 + \frac{1}{2} - \frac{1 + C4}{2} C11 + \frac{C4}{2} \sqrt{C6} + \frac{C8}{2} C14\right).$$

Expression (B.44) shows that C^* is independent on L, which is shown graphically in Fig. 5.

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